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Rapid-annealing effect on the microstructure and magnetic properties of the Fe-rich nanocomposite magnets

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The rapid-annealing effect on the microstructure and magnetic properties of the nanocomposite $Fe_{93-x-y}Co_xNb_2(Nd,Pr)_yB_5$ ($x=0-20$, $y=5-7$ at. %) alloys produced by crystallization of an amorphous phase have been investigated. The melt-spun ribbons consist of an amorphous phase in the as-quenched state and the amorphous phase changes to a nanocomposite structure consisting mainly of bcc-(Fe, Co), $(Nd,Pr)_2(Fe,Co)_{14}B$ and residual amorphous phases after annealing at temperatures of 973–1023 K. The nanocomposite alloys exhibit improved values of remanence (J_r), coercive force (H_{cJ}), and maximum energy product $[(BH)_{max}]$ upon annealing at a higher heating rate (α) in the temperature range corresponding to the primary crystallization temperature of bcc-Fe phase. As α increases, the grain sizes of each phase decrease, especially for the $Nd_2Fe_{14}B$ phase, and the ratio of total surface area of the $Nd_2Fe_{14}B$ to bcc-Fe phases (S_{hard}/S_{soft}), which are evaluated assuming each grain is a sphere, becomes close to 1 for the $Fe_{88}Nb_2Nd_5B_5$ alloy. This result indicates that the homogeneity of soft and hard magnetic phases is improved by rapid annealing, and it causes the improvement of hard magnetic properties of the Fe-rich nanocomposite alloys. [S0021-8979(00)23908-X]

I. INTRODUCTION

It has been reported that a nanocomposite alloy consisting of soft and hard magnetic phases behaves as a magnet consisting of a single hard magnetic phase,[1] and has the potentiality showing high $(BH)_{max}$.[2] Therefore, a lot of studies of the nanocomposite magnets have been made.[3–7] However, the nanocomposite magnets obtained in experiments until today show inferior hard magnetic properties to the value expected in theoretical simulations.

Schrefl *et al.* have reported[8] that the hard magnetic properties of the bcc-Fe/$Nd_2Fe_{14}B$ nanocomposite magnets significantly depend on the grain size and volume fraction of the bcc-Fe phase. The microstructure of the nanocomposite alloy formed by crystallization of an amorphous phase is sensitive to the annealing process, so that the optimum annealing is necessary for their good hard magnetic properties.

We have already reported[6,7] that the amorphous Fe-rich Fe–(Nb, Zr)–(Nd, Pr)–B melt-spun ribbons change to a nanocomposite structure of bcc-Fe and $(Nd,Pr)_2Fe_{14}B$ phases with the remaining amorphous phase in a boundary and show $(BH)_{max}$ of 60–110 kJ/m^3 after annealing at 970–1020 K. In this study, we report the rapid-annealing effect on the microstructure and magnetic properties of the Fe-rich Fe–(Nb, Zr)–(Nd, Pr)–B nanocomposite magnets produced by crystallization of an amorphous phase.

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II. EXPERIMENTAL PROCEDURE

Alloy ingots with composition of $Fe_{93-x-y}Co_xNb_2(Nd,Pr)_yB_5$ [$x=0-20$, $y=5-7$ (at. %)] were prepared by an arc-melting method. Amorphous ribbons 20 μm thick and approximately 1 mm broad were produced in an Ar atmosphere using a single roller melt spinning equipment with a copper wheel rotating with a velocity of about 42 m/s. The annealing treatment was performed for 180 s at temperatures (T_a) ranging from 923 to 1073 K with heating rates (α) between 0.05 and 3 K/s in a highly evacuated infrared furnace.

Structure was investigated by x-ray diffractometry and a transmission electron microscope (TEM), and the crystallization behavior was examined by differential scanning calorimeter (DSC). Magnetic properties were measured with a vibrating sample magnetometer (VSM) by applying a magnetic field of 1.5 T to the longitudinal direction of the sample.

III. RESULTS AND DISCUSSION

First, magnetic properties of the melt-spun ribbons annealed at various temperatures (T_a) with constant heating rates (α) of 0.05–3 K/s were investigated. The highest maximum energy product $[(BH)_{max}]$ is obtained at $T_a=973-1023$ K for each samples, and the optimum annealing temperature is unchanged with changing α. Figure 1 shows the remanence (J_r), coercive force (H_{cJ}), and $(BH)_{max}$ dependence on α for the $Fe_{88}Nb_2Nd_5B_5$, $Fe_{73}Co_{15}Nb_2Nd_5B_5$, $Fe_{76}Co_{10}Nb_2Nd_7B_5$, and $Fe_{66}Co_{20}Nb_2Pr_7B_5$ alloys annealed

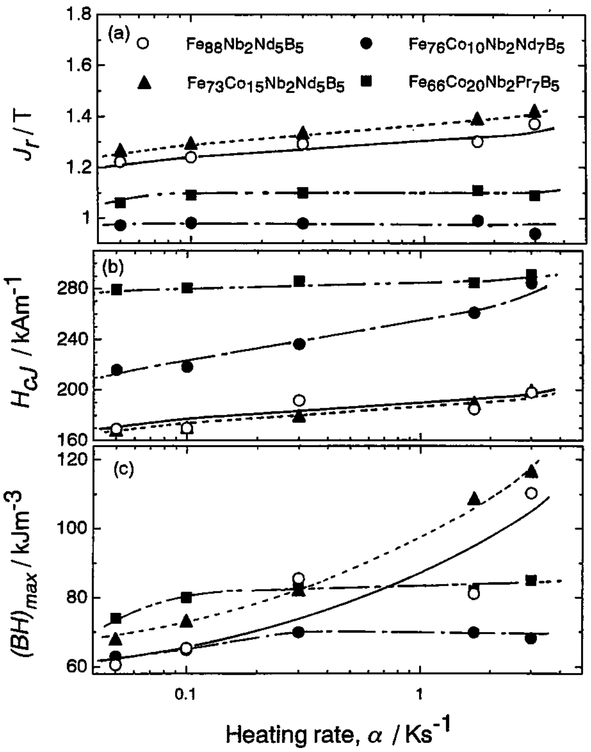

FIG. 1. Changes in the remanence (J_r), coercive force (H_{cJ}), and maximum energy product [$(BH)_{max}$] as a function of heating rate (α) for the $Fe_{88}Nb_2Nd_5B_5$, $Fe_{73}Co_{15}Nb_2Nd_5B_5$, $Fe_{76}Co_{10}Nb_2Nd_7B_5$, and $Fe_{66}Co_{20}Nb_2Pr_7B_5$ alloys annealed at the optimum temperature with constant α values between 0.05 and 3 K/s.

at the optimum temperature. It is noticed that J_r, H_{cJ}, and $(BH)_{max}$ become higher as α increases, and the increase in $(BH)_{max}$ is remarkable for the $Fe_{88}Nb_2Nd_5B_5$ and $Fe_{73}Co_{15}Nb_2Nd_5B_5$ alloys, which contain a high Fe, Co concentration of 88 at. %. This result indicates that rapid annealing improves the hard magnetic properties for the Fe-rich nanocomposite alloys. We chose the $Fe_{88}Nb_2Nd_5B_5$ alloy as a representative composition, and investigated the microstructure of the alloy annealed at an optimum temperature of 1023 K.

Figure 2 shows the DSC curves of the $Fe_{88}Nb_2Nd_5B_5$ amorphous alloy at a scanning rates of 0.10–0.67 K/s. Two exothermic peaks are seen at 830–870 and 930–960 K. X-ray diffraction analyses indicate that the first and second peaks are due to the precipitation of the bcc-Fe and $Nd_2Fe_{14}B$ phases, respectively. This result suggests that the alloy forms a composite structure of the bcc-Fe and $Nd_2Fe_{14}B$ phases after annealing at 1023 K.

The grain sizes of the bcc-Fe and $Nd_2Fe_{14}B$ phases are evaluated by using Scherrer's equation from the half width of bcc (110) diffraction peak and TEM observation. Figure 3 shows the change in the mean grain sizes as a function of α for the $Fe_{88}Nb_2Nd_5B_5$ alloy annealed at 1023 K. As α increases from 0.05 to 3 K/s, the mean grain sizes of the bcc-Fe and $Nd_2Fe_{14}B$ phases obtained by TEM observation decrease by 10% and 45%, respectively. It is noticed that the grain size reduction is remarkable for the $Nd_2Fe_{14}B$ phase.

Figure 4 shows the change of the magnetization as a function of temperature (the $J-T$ curve) for the

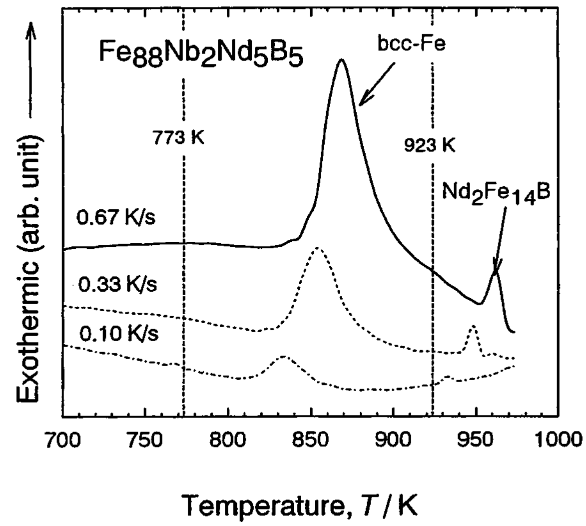

FIG. 2. DSC curves of the $Fe_{88}Nb_2Nd_5B_5$ amorphous alloy at scanning rates of 0.10–0.67 K/s.

$Fe_{88}Nb_2Nd_5B_5$ alloy annealed at $T_a = 1023$ K with $\alpha = 0.05-3$ K/s. We can see that the magnetization decreases with the increase of temperature, inflects near 600 K, and decreases with further increasing temperature. The inflection point is correspond to the Curie temperature of the $Nd_2Fe_{14}B$ phase (588 K), so that the magnetization in the temperature range above 600 K is due to the bcc-Fe phase. We can estimate roughly the volume fraction of the bcc-Fe phase by fitting the $J-T$ curve of pure Fe on the curve above the inflection point and that of the $Nd_2Fe_{14}B$ phase from the residual magnetization obtained by subtracting that of bcc-Fe phase because the magnetization is almost saturated for the

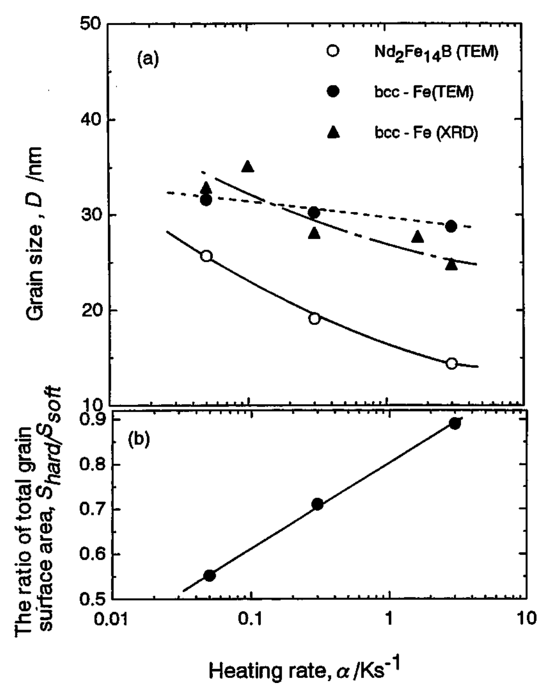

FIG. 3. (a) Changes in the mean grain size (D) and (b) the ratio of total surface area of the $Nd_2Fe_{14}B$ to bcc-Fe phases (S_{hard}/S_{soft}) as a function of heating rate (α) for the $Fe_{88}Nb_2Nd_5B_5$ alloy annealed at 1023 K.

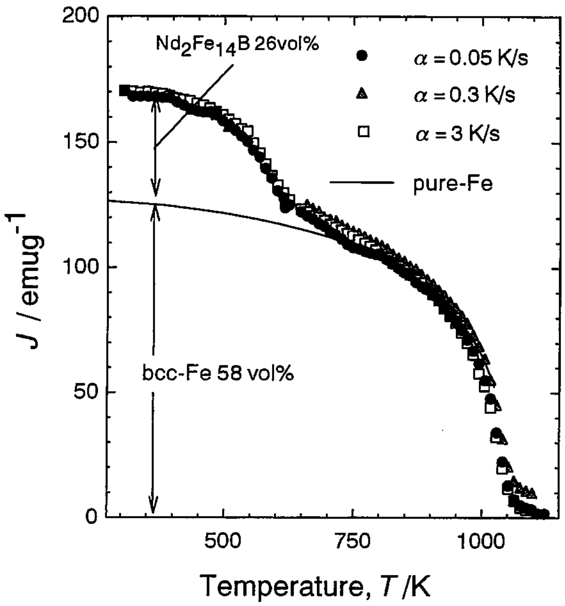

FIG. 4. Changes of the magnetization as a function of temperature for the $Fe_{88}Nb_2Nd_5B_5$ alloy annealed at temperature (T_a) of 1023 K with heating rates (α) of 0.05–3 K/s.

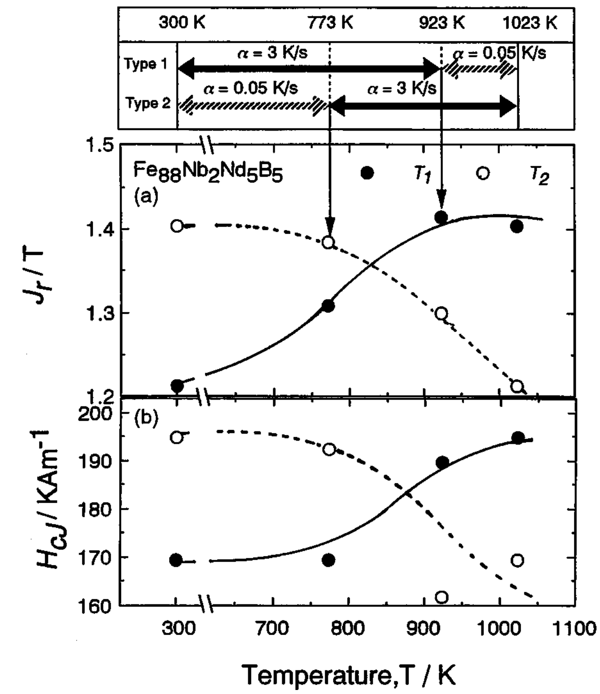

FIG. 5. Magnetic properties of the $Fe_{88}Nb_2Nd_5B_5$ alloy annealed with two types of conditions, one is to heat with heating rates (α) of 3 and 0.05 K/s for the temperature range from room temperature to T_1 and from T_1 to 1023 K, respectively, and the other is to heat with α of 0.05 and 3 K/s from room temperature to T_2 and from T_2 to 1023 K, respectively.

$Fe_{88}Nb_2Nd_5B_5$ alloy. The volume fractions of the bcc-Fe and $Nd_2Fe_{14}B$ phases are approximately 58% and 26%, respectively, and the total value is 84% not 100%, presumably due to the existence of the residual amorphous phase with low magnetization. The volume fractions of each phase are almost unchanged by changing α form 0.05 to 3 K/s.

Assuming that each grain is spherical, the ratio of total surface area of the $Nd_2Fe_{14}B$ to bcc-Fe phases (S_{hard}/S_{soft}) is calculated by using the grain size and volume fraction. The S_{hard}/S_{soft} value is shown in Fig. 3(b) as a function of α. As α increases, the S_{soft}/S_{hard} value increases and becomes close to 1, indicating that the probability that the $Nd_2Fe_{14}B$ grains adjoin the bcc-Fe grains becomes larger with increasing α. This result implies that rapid annealing improves the homogeneity of the soft and hard magnetic phase by the refinement of the hard magnetic phase comprising a small volume fraction. The improvement of hard magnetic properties caused by rapid annealing is presumably due to the increase of the exchange-coupled region between the soft and hard magnetic phases.

Next, two types of annealing with a bent heating rate were carried out for the $Fe_{88}Nb_2Nd_5B_5$ amorphous alloy. One is to heat the sample with heating rates of 3 and 0.05 K/s for the temperature range from room temperature to T_1 and from T_1 to 1023 K, respectively, and keep at 1023 K for 180 s (type 1). The other is to heat the sample with heating rates of 0.05 and 3 K/s from room temperature to T_2 and from T_2 to 1023 K, respectively, and keep at 1023 K for 180 s (type 2).

Figure 5 shows the dependence of T_1 and T_2 on the hard magnetic properties of the $Fe_{88}Nb_2Nd_5B_5$ alloy annealed with these conditions. J_r and H_{cJ} show high values in the temperature range above 923 K for T_1 and below 773 K for T_2, implying that rapid annealing is effective on applying in the temperature range between 773 and 923 K to improve the hard magnetic properties. We can notice that this temperature range is corresponding to the precipitation temperature of the bcc-Fe phase as shown in Fig. 2. These results indicate that rapid annealing in the precipitation temperature of the primary bcc-Fe phase causes the refinement of not only the bcc-Fe but also $Nd_2Fe_{14}B$ phases which precipitate subsequently around the bcc-Fe phase.[7] This is an interesting consequence and further investigations are required to clarify the refinement mechanism of the $Nd_2Fe_{14}B$ phase.

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